Electron-Phonon Coupling at the Tungsten Surface

Hydrogen Adsorbate Vibrations Split Surface Electron Band

For metals at very low temperatures, exotic states (e.g., superconductivity, charge-density waves, and spin-density waves) driven by "many-body" effects replace the ground (lowest energy) state predicted by conventional "one-electron" theory. Since many-body effects are often more prominent in low-dimensional metallic systems, such as two-dimensional surfaces and interfaces, researchers from the ALS and the University of Oregon have engaged in a continuing series of experiments to explore metal surfaces covered with one atomic layer (monolayer) or less of adsorbate atoms. In their most recent investigation of tungsten covered with one monolayer of hydrogen, the ALS/Oregon team has demonstrated a many-body effect in which there is a strong coupling between the tungsten surface electrons and vibrating hydrogen atoms that splits a surface energy band.

Metals at room temperature are traditionally described by the one-electron approximation in which the electrons act independently and have a spectrum of energies and momenta that form energy bands. Among metals, tungsten has a particularly long history as a subject for surface science and therefore provides a well-characterized starting point for the investigation of many-body effects at surfaces by the technique of angle-resolved photoemission (ARPES). With ARPES, researchers can create energy-momentum "maps" of the energy bands for both "bulk" electrons with momenta in three dimensions and surface electrons with momenta in two dimensions parallel to the surface.

In earlier ARPES studies, the ALS/Oregon team discovered that the momenta of electrons at the Fermi energy for two-dimensional energy bands on the (110) surface of tungsten were highly sensitive to the amount of hydrogen on the surfaces. It is one of these surface bands that the new ARPES measurements show is altered by the hydrogen atomic vibrations. In the one-electron model, the creation and destruction of lattice-vibrational quanta (phonons) is a principal mechanism for driving transitions between quantum states as electrons relax or are thermally excited. But in the new experiments, the connection between lattice vibrations and electrons is much more intimate, and it is necessary to think of the two entities forming a composite, many-body system with its own spectrum of energies.

The tell-tale sign of the electron-phonon interactions involving hydrogen is that the energy for the onset of the band splitting matches the energy of the symmetric hydrogen stretching vibration mode. The electron vacancy (hole) left as a result of photoemission has two possible characters depending on its energy. If the hole energy is sufficiently large, its lifetime is short because it can decay into phonons, whereas holes with energies closer to the Fermi energy cannot decay into real phonons. Instead, these holes are longer-lived and carry a cloud of "virtual" phonons that effectively raise the mass of the hole. The hydrogen vibration frequency sets the energy scale that divides these two characteristic behaviors.

The critical test of this explanation is that substituting deuterium (which is heavier than hydrogen and consequently has a smaller vibrational frequency) for hydrogen decreased the onset energy for band splitting. Moreover, among the various surface and bulk bands present, only the particular surface band that is most strongly confined to the surface shows a strong electron-phonon coupling effect, evidence that the interaction is confined to the hydrogen surface layer.

The results have potential practical implications. That electrons and holes can decay to phonons also implies the reverse: that phonons can decay by excitation of electron-hole pairs. This is an important dissipation channel for energy and will affect chemical reactions at surfaces, such as reactions on catalysts. Another interesting implication is the possibility of an electron-phonon-mediated superconductivity that is confined to the surfaces of metals and has a greatly enhanced critical temperature as compared to ordinary superconductors. ■

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E. Rotenberg, J. Schaefer, and S.D. Kevan "Coupling between adsorbate vibrations and an electronic surface state," *Phys. Rev. Lett.* **84**, 2925 (2000).

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- **Many-body effects at the frontier of condensed-matter science**
  - *Metallic ground state at low tempertures differs from one-electron predictions*
  - *Superconductivity, charge-density waves, spin-density waves are examples*
  - *Often more prominent in low-dimensional systems such as surfaces*
- **Tungsten surface a model two-dimensional system**
  - *Geometrically and electronically well characterized over many years*
  - *The surface is dominated by two-dimensional electronic states*
  - *These states can be easily perturbed, but not killed, by depositing overlayers*
  - *Angle-resolved photoemission (ARPES) yields surface band structure*
- **ARPES study of tungsten surface at the Advanced Light Source**
  - *Continuing series of ALS/University of Oregon experiments*
  - *Explore many-body effects in two dimensions*
  - *Early suggestion that surface bands sensitive to adsorbed hydrogen*
- **Effect of one monolayer of hydrogen on surface electrons**
  - *Surface-localized band split by electron-phonon interaction*
  - *Verified by deuterium substitution and lack of splitting in deeper bands*
  - *Possible influence on surface chemistry, catalysis*

E. Rotenberg (ALS) and J. Schaefer and S.D. Kevan (University of Oregon). [*Phys. Rev. Lett.* **84**, 2925 (2000]

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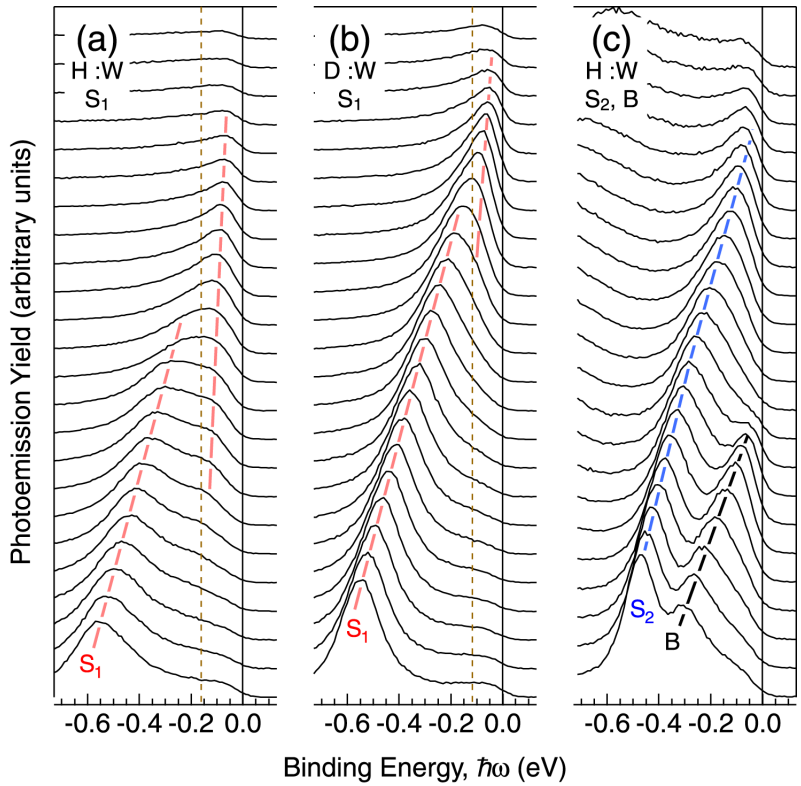

(a) For hydrogen on tungsten(110), the splitting of the S1 surface band near the energy of the symmetric hydrogen stretching mode (dashed line) is clearly visible in ARPES spectra. (b) On substituting deuterium for hydrogen, the vibrational energy decreases because of the H-D isotope effect, and the splitting becomes smaller. (c) In less surface-localized bands (S2 and B) having poorer overlap to the surface phonon modes, the effects for H disappear altogether.

E. Rotenberg (ALS) and J. Schaefer and S.D. Kevan (University of Oregon). [*Phys. Rev. Lett.* **84**, 2925 (2000]

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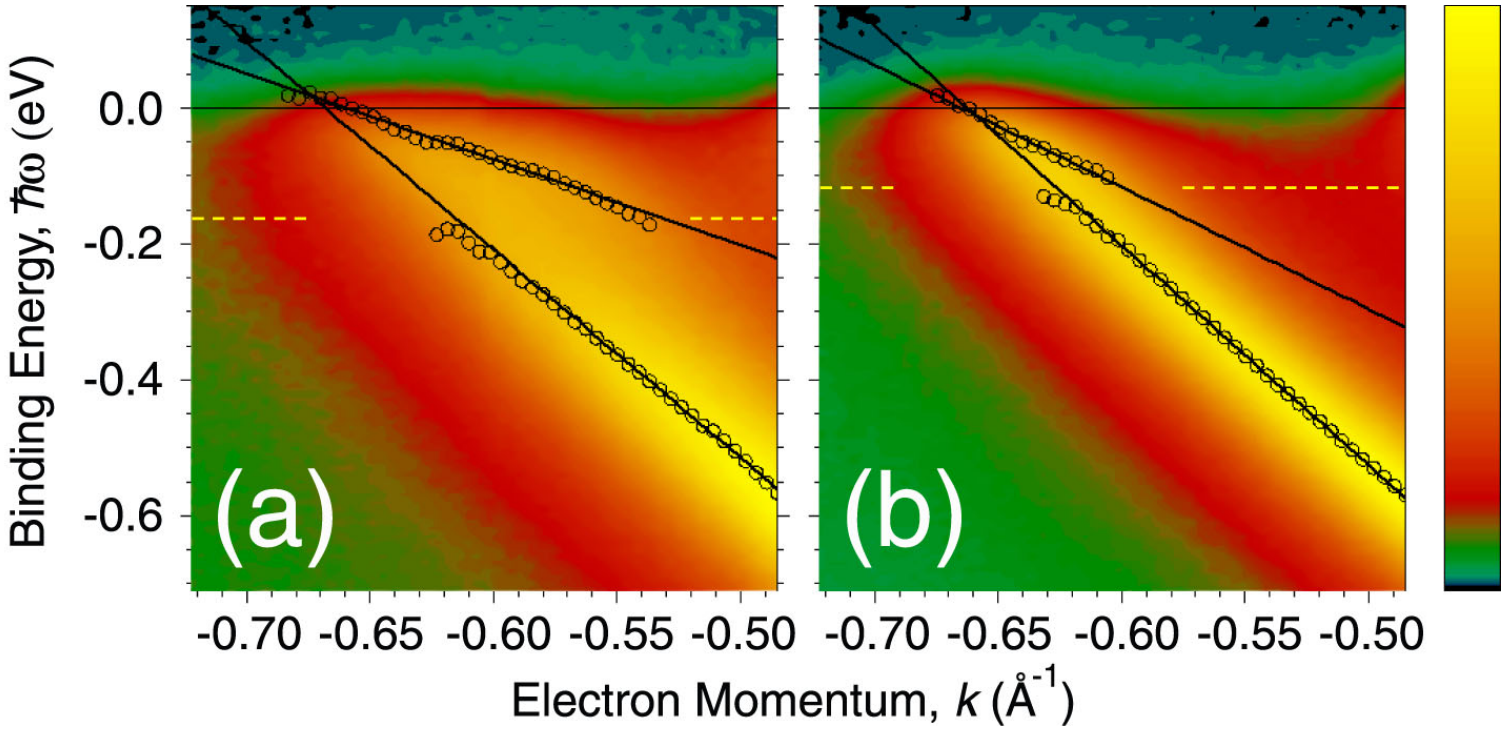

ARPES intensity maps of surface band S1 in the presence of (a) hydrogen and (b) deuterium with the fitted positions (circles) of the two peaks, the best-fit lines through these data, and the known hydrogen and deuterium symmetric stretch vibrational energies (horizontal dashed lines). From the slopes of these lines, one can derive the electron-phonon coupling parameters.

E. Rotenberg (ALS) and J. Schaefer and S.D. Kevan (University of Oregon). [*Phys. Rev. Lett.* **84**, 2925 (2000]